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The formation of the charged exciton complexes in self-assembled InAs single quantum dots

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Abstract. We study the low-temperature photoluminescence (PL) of the self-assembled InAs single quantum dots (QDs) using conventional μ-PL setup to detect PL from an individual QD. We demonstrate, that at certain experimental conditions, what concerns the laser excitation energy, the laser power and the sample temperature, several novel lines appear in the PL spectra. The lines, redshifted relative to the exciton PL line, are interpreted in terms of charged exciton complexes.

Introduction

Multiparticle effects in quantum dots (QDs), which were studied both theoretically [1, 3] and experimentally [2–5], determine the performance of a number of optoelectronic devices, for example QD lasers and single-electron transistors, which in turn are considerably affected by the charge, stored in the QD [6]. This highlights the outstanding role of multiparticle complexes with non-equal number of electrons e and holes h which were spectroscopically examined on large ensembles of QDs [2, 3] as well as in single QDs [4, 5]. In the latter type of studies, the effect of inhomogeneity is avoided but is still associated with some disadvantages: The supplying of extra electrons into the QD from the doped layers [4] is based on the use of an external electric field, which in turn affects the entire spectrum of the QD. Feeding the QD with the extra charges from the impurity atoms positioned in a close vicinity of a QD [5] makes the transfer of excess carriers into QD random and hardly controlled.

In the present contribution we propose a new method to create charged excitons in QDs. This is based on the possibility to monitor the photogenerated carrier diffusivity by means of tuning the laser excitation energy ($h\nu$) and changing the crystal temperature (T). As a result, under certain experimental conditions, we can detect the appearance of two new lines in photoluminescence (PL), redshifted relative to the fundamental single exciton line. These novel lines are interpreted in terms of charged exciton complexes.

1. Samples and experimental setup

The sample studied consists of one layer of InAs self-assembled QDs between two 100 nm wide GaAs barriers (the MBE growth procedure used is described in detail elsewhere [7]). The QDs formed are lens shaped with a lateral size of 35 nm and a height of 10 nm. Typical interdot distance is 10 μm, which allows the use of a conventional micro-PL setup (described in detail in Ref. [8]) in order to spatially resolve individual dots. The laser beam could be focused on the sample surface with a spot size down to 2 μm, to excite and study

the PL from a single QD. The cw light of a Ti-Sp laser was used for the photoexcitation. The sample temperature was maintained in the range 4 K $\leq T \leq$ 40 K.

Four single quantum dots located at different spatial positions of the sample were examined in this study. All of them revealed an analogous behavior and for consistency, we present the results taken from only one single quantum dot to demonstrate a typical behavior of the QDs.

2. Experimental results and discussion

Figure 1 shows PL spectra measured at $T = 4$ K on a selected single QD taken for a wide range of excitation powers, P_{ex}, and excitation energies, $h\nu$. At lowest P_{ex} and $h\nu = 1.684$ eV (Fig. 1(a)), the spectrum consists of a single line, X. With increasing P_{ex}, two groups of lines appear in the spectra: One is redshifted relatively the X line (by 3–7 meV), while the other (marked as X_{pp}) appears blueshifted by a 30 meV. At highest P_{ex}, the intensity for the first group of lines tends to saturate and even decrease, while the intensity of the second one progressively increases. These observations imply that the first (second) group of lines originates from the ground (first excited) state of the QD. For another excitation energy $h\nu = 1.536$ eV (Fig. 1(b)), a qualitatively similar behavior for the PL spectra was revealed at high P_{ex}, while the spectrum taken at lowest P_{ex} differs markedly from the corresponding one in Fig. 1(a). Two new lines, marked as X^- and X^{--}, appear in this spectrum.

We will below propose an explanation for these dramatic changes obtained at lowest P_{ex} initiated by the tuning of $h\nu$. We first note that as this difference occurs at lowest P_{ex}, we can exclude the occupation with more than one exciton ($e - h$ pair) in the QD as an explanation. Second, the appearance of X^- and X^{--} when changing $h\nu$ can not be explained in terms of impurity related transitions, because these would be observed at any $h\nu$ [5]. Instead, we exploit a model which predicts a dependence of the photogenerated free carrier diffusivity on $h\nu$. Indeed, carriers being excited in the GaAs barriers (Fig. 1(a) and (b)) can release their kinetic energy either via fast emission of LO phonons (with energy $\hbar\omega_{LO}$), or by an emission of a cascade of acoustic phonons, which takes much longer time.

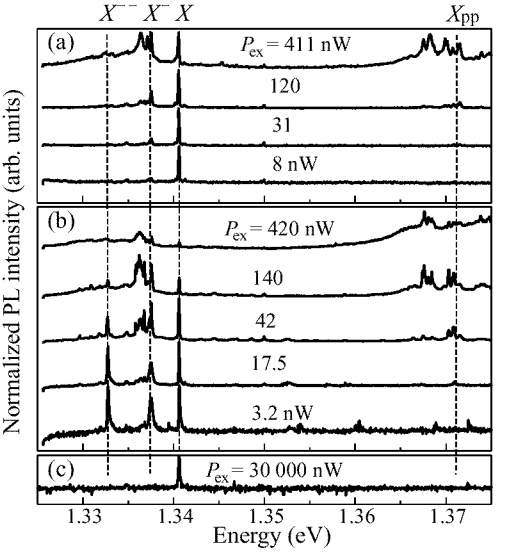

Fig. 1. Normalized PL spectra of an individual QD taken at $T = 4$ K, for a number of P_{ex} at different pump-photon energies:
(a) $h\nu = 1.684$ eV,
(b) $h\nu = 1.536$ eV
and (c) $h\nu = 1.433$ eV.

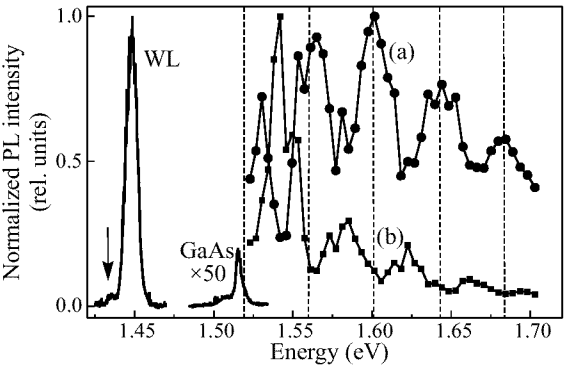

Fig. 2. The normalized peak PL intensities of (a) X and (b) X^{--} lines as a function of $h\nu$, taken at $T = 4$ K and $P_{ex} = 10$ nW. The normalized PL spectrum of the wetting layer (WL) and of the GaAs barrier is also shown for the same values of T and P_{ex} at $h\nu = 1.536$ eV.

As a result we expect that carriers, for which the kinetic energy fits with an integer number of $\hbar\omega_{LO}$, become motionless at the band edge shortly after generation, while for any other kinetic energy, a much longer time for carriers is expected to cool down. Consequently this carrier is able to move a much longer distance in the crystal. Eventually, the latter case results in a higher diffusivity, which in turn increases the probability for this carrier to pass in the close vicinity of a QD and to become captured by the dot.

We consider the electrons as more likely candidates for such a process, since electrons in GaAs are seven times lighter than the holes. Thus we explain the appearance of the two new lines X^{-} (X^{--}) at a certain $h\nu$ as a recombination of complexes consisting of a single exciton and one (two) extra electron(s) captured to the QD to form negatively charged excitons. This assignment is supported by the prediction [1] that only electrons can be bound to the exciton in lens shaped QDs, as well as by the comparison of the binding energies for X^{-} (X^{--}) derived from experiment as the differences in the PL energy positions between X and X^{-} (X^{-} and X^{--}) lines as 3.1 meV (4.7 meV) with calculated ones of 2.6 meV (4.9 meV) using simple perturbation theory [3]. If one excites only immobile carriers, e.g. by excitation resonant with the localized states of the WL (as marked by an arrow in Fig. 2), no lines except X are expected in the PL spectrum. This is in full agreement with our experiment (Fig. 1(c)).

As follows from our model, the electron diffusivity should be modulated with $h\nu$, and accordingly, the amplitudes of the neutral and charged exciton lines should oscillate. In fact such a behavior is nicely demonstrated in Fig. 2, where the amplitudes of the line X (Fig. 2(a)) and X^{--} (Fig. 2(b)) are shown as a function of $h\nu$. They oscillate in counterphase as expected with a characteristic period of 41.4 meV (shown by the vertical dotted lines) which fits well with the calculated energy difference of $h\nu$ changing the electron part of the kinetic energy by the LO phonon energy.

An alternative way to increase the electron diffusivity, and hence to initiate the appearance (or enhance the intensity) of the charged exciton lines is to increase the sample temperature. The results are shown in Fig. 3, for three characteristic $h\nu$. For the excitation energy $h\nu$, which gives rise only to the X line at lowest T (Fig. 3(a)), the increase of T evidently redistributes the PL intensity in favour of X^{-} and X^{--} lines. For excitation energies $h\nu$ at which the three lines are of comparable intensities at low T (Fig. 3(b)), the X line evidently disappears at highest T and the spectrum is instead dominated by the X^{--} line. In the case of excitation of solely localized carriers at $h\nu = 1.433$ eV (at the arrow in

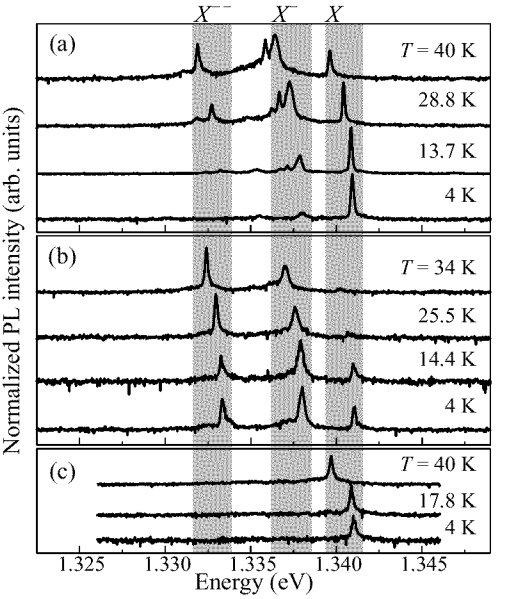

Fig. 3. Normalized PL spectra of a selected QD taken for a number of T and different excitation energies: (a) $h\nu = 1.686$ eV and $P_{ex} = 5$ nW, (b) $h\nu = 1.557$ eV and $P_{ex} = 5$ nW and (c) $h\nu = 1.433$ eV and $P_{ex} = 30000$ nW.

Fig. 2), no diffusion of carriers is expected and in the PL spectra only a small redshift of X is observed at higher temperatures (Fig. 3(c)) due to the band gap shrinkage (the same as in Fig. 3(a) and (b)). This result (Fig. 3(c)) confirms that the temperature effect on the PL spectra is due to an increased photogenerated carrier diffusivity and not due to an activation of localized electrons (for example electrons localized at impurities in the close vicinity of the QD).

Thus, the present study demonstrates a new method to create charged excitons by changing the effective carrier diffusivity by means of an altered excitation energy and/or sample temperature.

Acknowledgements

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